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Integrated process of fungal membrane bioreactor and photocatalytic membrane reactor for the treatment of industrial textile wastewater

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ABSTRACT

In this study, fungal membrane bioreactor (FMBR) and semiconductor photocatalytic membrane reactor (PMR) were used in order to test the efficiency of integrated fungal biodegradation and photocatalytic degradation of textile wastewater from reactive washing processes. It was found that color removal and chemical oxygen demand (COD) reduction efficiencies were 88% and 53% for photocatalytic degradation, respectively. TiO_2 and ZnO were tested as semiconductor catalysts in the PMR and TiO_2 showed better efficiencies than ZnO for both color and COD removal. However, it was attained that color removal and COD reduction efficiencies were about 56% and 60% for fungal biodegradation using *Phanerochaete chrysosporium*, respectively. Moreover, integrated system in which photocatalytic degradation was employed as a post-treatment application after fungal biodegradation process achieved high removal efficiencies for color and COD removal as 93% and 99%, respectively.

1. Introduction

Water is an essential substance in virtually each industry especially for textile industry with direct and indirect utilization in the different processes (cooling water, chilled water, washing water etc.). Industry collects fresh water from water resources located near natural areas such as lake, river, underground etc. However, these resources are rapidly depleting [1]. The textile industries consume large amount of fresh water and produce large volumes of wastewater in different steps while dyeing and finishing processes. In textile industry, wastewater containing excessive color, COD, and complex chemicals is produced. These parameters are considered as critical contaminants. This kind of wastewater contains a large amount of dyes which are considerably toxic to living systems. Therefore, textile wastewater has been considered an urgent need for protection and providing aquatic life cycle.

Due to the fact that extremely changing the content of textile industry, its effluent contains complex chemicals. In this perspective, for effective decolorization of textile industry effluent requires some kind of fungal assistance and suitable reactor conditions. Certain textile effluent contains dyes, salts, caustic, surfactants chelating agents, by-products, precursors etc. [2].

Many processes which include physicochemical, biochemical, combined treatment processes and other advanced treatment technologies have been studied to treat textile wastewater up to now. However, the application of these processes in an industrial scale becomes difficult due to the operational problems, maintenance and costs. Chemical oxidation via ozone, or a combination of ozone and H_2O_2 has great interest but their costs are quite high. In addition, biological treatment by activated sludge allows high efficiency for COD removal, but it does not provide complete color removal and some potential problems such as bulking and foaming can occur [3].

Fungi which have broad range degradation capacity were found to be most efficient in breaking down synthetic dyes. White-rot fungi are the most promising biological agents which can be utilized for developing novel biological wastewater treatment processes such as fungal membrane bioreactor. They are able to degrade a wide range of recalcitrant pollutants including various types of dyes, lignocellulose compounds, organic pollutants such as poly aromatic hydrocarbons (PAHs), chlorophenols and poly chlorinated biphenyls. These microorganisms finally break down the compounds via their enzymatic reactions (such as manganese peroxidases, lignin peroxidases, laccase etc.) [4–6].

Blanquez et al. studied the operational conditions for the continuous treatment process of the metal complex dye in a fluidized

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bed bioreactor using air pulses with retained pellets of the white rot fungus, *Trametes versicolor*. It was reported that decolorization efficiency was calculated as >80% for the different hydraulic retention times (HRTs) ranging from 18 to 120 h, and the dye removal rates ranged from 1.16 to 6.73 mg/L/h [7]. Therefore, HRTs and biomass concentrations considered as significant parameters during decolorization of real textile effluents. Blanquez et al. also investigated the decolorization of textile wastewater using white rot fungus pellets, *T. versicolor* in the scale up of a 10 L air pulsed bioreactor. Decolorization experiments of 150 mg/L Grey Lanaset G dye solutions showed that in both discontinuous and continuous treatment with an HRT of 48 h, the decolorization efficiency was higher than 90% [8].

Photocatalytic processes for wastewater treatment are commonly studied in powder suspended and immobilized (fixed) semiconductor systems. Photocatalytic degradation efficiency is affected by many parameters such as; pH, temperature, type of semiconductors, pollutant concentration, contact time and illumination intensity [9–14]. Photocatalytic membrane reactor (PMR), in which photocatalysis is coupled with membrane separation process, is a new hybrid technology for water and wastewater treatment. With some unique advantages such as nontoxic and continuous running, PMR was developed rapidly in the past few years [15]. Moreover, a low-cost and eco-friendly recovery of the photocatalysts and the separation of products and/or inter mediates simultaneously occur in PMR. In the meantime photocatalytic process improves membrane cleaning and delays membrane fouling [16].

In the past decades, various hybrid processes were being developed for wastewater treatment. For example, Mozia et al., reported that the results of the investigation on the possibility of coupling photocatalysis and membrane distillation (MD) for degradation of acid red 18 in aqueous solutions [17]. In other study, the successful operation of a hybrid photocatalysis–membrane separation process (in a laboratory-scale pilot system) was demonstrated for degradation of humic acids (HA). The photocatalytic membrane reactor (PMR) employing an ultrafiltration (UF) submerged module was operated in continuous mode with TiO_2 catalyst [18].

In recent studies, fungal membrane bioreactor or photocatalytic membrane reactor processes have been investigated separately to treat different and complex wastewater effluents [19–28]. PMR allows the separation characteristics of membrane in order to separate the treated effluent and to maintain constant semiconductor suspension within the photoreactor. The membrane can act as a barrier to confine the catalyst within the system. Also, FMBR provides constant fungi suspension within the bioreactor. Moreover, the membrane keeps the enzyme released from fungi in the reactor.

The aim of this study is to develop a novel integrated treatment process involving fungal biodegradation in FMBR and ultraviolet assistedphotodegradation in PMR. Theseprocesses were used integrated in order to achieve high color and COD removal in industrial textile wash water effluent. The primary objective of this study was to utilize the integrated feature of organic matter degradation by white-rot fungi and color removal by semiconductor photocatalytic reduction sequentially. The integrated process performance was followed up in bench-scale reactors in sequential process to evaluate the long term performance of FMBR and PMR for high color and COD removal from washing bath wastewater.

2. Materials and methods

2.1. Materials

2.1.1. Chemicals

Commercially pure ZnO (particle size <1 μm, Sigma–Aldrich) and TiO_2 (particle size <1 μm, Evonik P-25) were used as semiconductor powders for PMR experiments. A flat plate membrane module with MP005 membrane type was used in both PMR and FMBR (Table 1). Textile wastewater was provided from a textile factory in Kayseri, Turkey. The characterization parameters of textile reactive washing bath wastewater were given in Table 2. The types of dyes that were utilized in this sort of textile industry were supplied by the company as follows; sunfron red, sunfix blue, sunfix deep red, sunfix red, sunfix navy blue, sunfix orange, sunfix yellow, sunzol navy blue, sunfron blue, sunzol turquoise blue, sunfix scarlet. The deionized water used in all the experiments was obtained from Milli-Q ultrapure water system.

Table 1
Properties of ultrafiltration membrane.

MP 005 Membrane	Properties
Membrane material	PES (polyethersulfone)
Manufacturer	Microdyn-Nadir GmbH
Membrane configuration	Flat sheet
Pore size	0.05 μm
Pure Water Flux [L/m^2/h]	>800
Module	Properties
Module configuration	Flat plate
Effective membrane area	30 cm^2
Module material	Teflon
Module size (W × L cm)	3 × 20

Table 2
Characteristic parameters of textile reactive washing bath wastewater.

Parameters	Value
Total COD (mg/L)	1125 ± 70
Soluble COD (mg/L)	800 ± 100
Suspended Solids (mg/L)	35 ± 2
Conductivity (μS/cm)	2660 ± 16
pH	8.7 ± 0.5
Color	Very dark violet

2.1.2. Potato Dextrose Agar (PDA)

Potato Dextrose Agar (PDA) was growth medium of *Phanerochaet echrysosporium* and it was purchased from Merck and prepared as indicated on the PDA package. This medium was autoclaved at 121 °C for 15 min. After cooling to atmospheric temperature (below 40 °C) the media was poured into the test tubes or Petri dishes for solidification in the laminar chamber and inoculated from the stock culture. Taking out from the chamber of these tubes or Petri dishes were incubated in a BOD incubator for fungi growth.

2.1.3. Fungus strain

Phanerochaete chrysosporium was obtained from Environmental Microbiology Laboratory of Environmental Engineering Department, University of Nigde, Turkey. It was white in color and had a mycelia structure. The fungus was available on PDA slant in a test tube.

2.2. Methods

2.2.1. Making sub-culture of fungus strain

P. chrysosporium was grown for 5 days at 30 °C in PDA plate and it was used to inoculate 250 mL erlenmeyer flasks with 50 mL of stock basal media which was prepared by glucose (2 g/L) and other nutrients into Milli-Q water. The other components of the stock basal media were as follows: 2 g/L peptone, 2 g/L KH_2PO_4, 0.1 g/L $CaCl_2$, 0.5 g/L $MgSO_4.7H_2O$, 0.001 g/L thiamine and 1 mL/L trace elements. Stock trace elements solution was prepared by dissolving 0.08 g $CuSO_4.5H_2O$, 0.05 g $NaMoO_4.2H_2O$, 0.07 g $MnSO_4.4H_2O$, 0.043 g $ZnSO_4$, 0.05 g $Fe(SO_4)_3$ in 1 L of ultrapure water. The solution